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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of:

Tomokuni, et al.

Serial No.: 09/976,036 Group Art Unit: 1773

Filed: October 15, 2001 Examiner: Ahmed, S

For: Fiber-Reinforced Plastic Molded Article, its Production Method and a Molding Mold Using that Method

DECLARATION UNDER 37 CFR §1.132

I, Hidehiko TOMOKUNI, hereby declare and state that:

1. I am a citizen of Japan, residing at 4-35 Izumi Ootsu-shi, Osaka, Japan.

2. I work in the section of Dainippon Ink and Chemicals, Inc., in which research and development related to the present invention were performed. I am one of the inventors of the present invention, and I am fully familiar with the subject matter of the present application as well as the references relied upon by the Examiner in the prosecution of this application.

3. I obtained a bachelor's degree from Tokai University, Faculty of Engineering, Department of Industrial Chemistry, in March 1990.

4. I am currently employed by Dainippon Ink and Chemicals,

Inc., and began working for Dainippon Ink and Chemicals, Inc., in April 1990, where I have engaged in research and development relating to unsaturated polyester resins.

5. I have conducted the tests described below.

Object of Tests

The purpose of the tests is to prove that tensile elongation percentage of the cured product of the unsaturated polyester resin composition disclosed in Kurtz et al. is less than 3%, which is the lower limit of tensile elongation percentage in the present invention.

Method

<Synthesis of Unsaturated Polyester Resin>

Based on the composition shown in Table II of Kurtz et al., the composition for the tests were prepared. Specifically, 1003.2g of propylene glycol (M.W. 76g/mol), 1302.4g of phthalic anhydride (M.W. 148g/mol), and 431.2g of maleic anhydride (M.W. 98g/mol) were charged into a 5L four-mouth flask equipped with a thermometer, stirrer, a reflux condenser, and a nitrogen feed tube. Under a nitrogen atmosphere, the temperature of the reaction mixture was raised to 205°C, by carefully not causing dehydration. The temperature was maintained at 205°C, until the solid acid value of 36.0 was obtained. Next, 0.1g of trihydroquinone was added to the flask and the

mixture was cooled to less than 140°C, and then 2045g of styrene monomers were added and dissolved thoroughly, thereby forming unsaturated polyester resin composition containing 45 wt% styrene monomer. The following resin constants at 25°C were obtained: 1.3 Pa·s of Viscosity, and 19.0 of Acid Value.

<Preparation of Casting Plate>

Two 35 cm X 35 cm glass plates with a thickness of 5 mm, of which a surface was subjected to release treatments, was prepared. The treated surfaces were faced to each other with rubber strips (width: 20 mm, thickness: 3.2 mm) interposed therebetween on three sides of the glass plates. The glass plates were fixed using clips, thereby forming a casting plate. 300g of the composition prepared according to the above-described method, 0.9 g of cobalt naphthenate, and 3.0 g of 55% methylethylketone peroxide were mixed, degassed under vacuum, and then charged into the casting plate. The mold was kept at room temperature, subjected to 2 hours of after-curing at 120°C, and the cured product was released from the casting plate.

<Tensile Elongation Test>

According to the method specified in JIS-K-7113, tensile elongation was measured (test speed of 5 mm/min, n=5).

<Result>

	n=1	n=2	n=3	n=4	n=5	Average
Tensile Strength (MPa)	58	62	58	59	60	59
Modulus of Tensile Elasticity (GPa)	4.3	4.5	4.4	4.4	4.4	4.4
Elongation Percent (%)	1.3	1.6	1.4	1.5	1.6	1.5

<Conclusion>

The elongation percents obtained for the composition were all below 3.0 % which is the lower limit in the present invention.

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: May 28, 2004

Hidehiko Tomokuni

Hidehiko TOMOKUNI

English translation of two document excerpts regarding tensile elongation percentage of general unsaturated polyester resins

(1) "PRACTICAL PLASTIC ENCYCLOPEDIA" page 230, first edition, published by SANGYO CHOSAKAI, on May 1, 1993

Table 2-9

Properties of FRP made of unsaturated polyester resin by each forming process

Items	Hand lay up	Spray up	MMD*1 product	Drawn*2 product	FW product
...					
...					
Tensile elongation percentage (%)	1.0-1.5	1.0-1.5	1.0-1.5	1.6-2.5	1.6-2.8
...					
...					

*1... MMD: Matched metal die

*2... FW: Filament winding (helical winding)

(2) "POLYESTER RESIN HANDBOOK" page 270, first edition, published by The Nikkan Kogyo Shimbun, Ltd., on June 30, 1985

Fig. 7.17* Unsaturated percentage of propylene glycol / phthalic anhydride / maleic anhydride-based resin, and elongation percentage according to the amount of styrene

Y-axis: Elongation percentage (%)

X-axis: Maleic anhydride (mol %)

--○-- Styrene 20%

--×-- Styrene 30%

--●-- Styrene 40%

--Δ-- Styrene 50%

*This graph includes the mole ratio of the units defined in claim 12 of Kurtz.

The mole ratio of the unit defined in claim 12 of Kurtz is as follows.
Ortho-phthalic acid : maleic acid : propylene glycol = 2 : 1 : 3 (mol)

The mol % of maleic acid to the whole is 17 mol% (= 1/6).

On the other hand, in Fig. 7.17, when maleic anhydride is 20 mol%, even if styrene content is 50%, the elongation percentage is no more than 1 %. Accordingly, when maleic acid is 17 mol%, the elongation percentage thereof would be obviously no more than 3%.

表２－８　ＦＲＰの機械的性質

項目	試験方法	単位	不飽和ポリエステル樹脂硬化物の特性				
			オルト系	イソ系	テレ系	ビス系	ハロゲン系
ガラス含有率	－	%	30	30	30	30	30
バーコール硬さ（#934）	JIS K 6919	－	40～55	45～60	45～60	45～55	45～60
曲げ強さ	JIS K 6919	MPa	180～210	190～220	190～220	180～200	180～200
曲げ弾性率	JIS K 6919	GPa	6.90～8.50	7.00～8.50	7.00～8.50	6.50～8.00	7.00～8.00
引張強さ	JIS K 6919	MPa	110～130	120～140	120～140	110～130	110～100
引張弾性率	JIS K 7113	GPa	6.90～9.00	7.00～9.00	7.00～9.00	6.80～8.50	7.00～9.00
圧縮強さ	JIS K 6919	MPa	130～180	130～180	130～180	120～170	120～170
シャルピー衝撃強さ	JIS K 6911	J/cm	6.0～8.0	7.0～9.0	7.0～9.0	6.0～8.0	6.0～8.0

項目	試験方法	単位	ビニルエステル樹脂		
			汎用	耐熱	難燃
ガラス含有率	－	%	30	30	30
バーコール硬さ（#934）	JIS K 6919	－	40～50	45～55	45～55
曲げ強さ	JIS K 6919	MPa	180～210	180～210	180～210
曲げ弾性率	JIS K 6919	GPa	7.50～9.00	7.50～9.00	7.00～8.50
引張強さ	JIS K 6919	MPa	120～140	120～140	110～130
引張弾性率	JIS K 7113	GPa	7.50～9.00	7.50～9.00	7.00～8.50
圧縮強さ	JIS K 6919	MPa	130～180	130～180	－
シャルピー衝撃強さ	JIS K 6911	J/cm	8.0～10.0	7.0～8.0	7.0～8.0

FRPの成形条件　(1)ガラス構成：#450，チョップドストランドマット3層
(2)硬化剤　：メチルエチルケトンペルオキシド，1.0wt%
(3)硬化条件　：室温ゲル化後，50℃×3hアフターキュア

表２－９　成形法別の不飽和ポリエスル樹脂ＦＲＰの特性

項目	ハンドレーアップ	スプレーアップ	MMD成形品	引抜成形品	FW成形品
ガラス基材	チョップマット	チョップ	チョプ（マット）	ロービング	ロービング
ガラス含有率（wt%）	30～40	30～40	30～50	50～80	60～90
比　重	1.4～1.8	1.4～1.6	1.5～1.7	1.6～2.2	1.7～2.3
引張強さ（MPa）	70～140	60～130	70～170	560～1300	560～1800
引張弾性率（GPa）	5.60～12.70	5.60～12.00	5.60～12.70	28.00～42.00	28.00～63.00
引張伸び率（%）	1.0～1.5	1.0～1.5	1.0～1.5	1.6～2.5	1.6～2.8
圧縮強さ（MPa）	110～180	110～180	130～210	210～490	350～500
曲げ強さ（MPa）	140～280	110～220	180～320	700～1300	700～1900
曲げ弾性率（GPa）	8.00～13.00	7.00～8.50	8.80～13.00	28.00～49.00	35.00～49.00
衝撃強さ（ft･lb/in）	5～25	5～15	10～20	45～60	40～60
硬さ（ロックウェル）（H）	40～105	40～105	40～105	80～110	M 98～120
熱伝導率（kcal/m･h･℃）	0.16～0.23	0.15～0.18	0.16～0.22	0.24～0.28	0.24～0.28
比　熱（kcal/kg･℃）	0.30～0.33	0.31～0.34	0.30～0.33	0.23～0.25	0.23～0.25
線膨張率（10^{-6} /℃）	18～32	22～36	18～32	5～14	4～11
熱変形温度（℃）	180～200	180～200	180～200	160～190	180～200
常用温度限界（℃）	65～160	65～160	65～160	65～160	100～200
絶縁耐力（V/mil）	200～400	200～400	200～400	200～400	200～400

MMD：マッチドメタルダイ，FW：フィラメントワイディング（ヘリカル巻）

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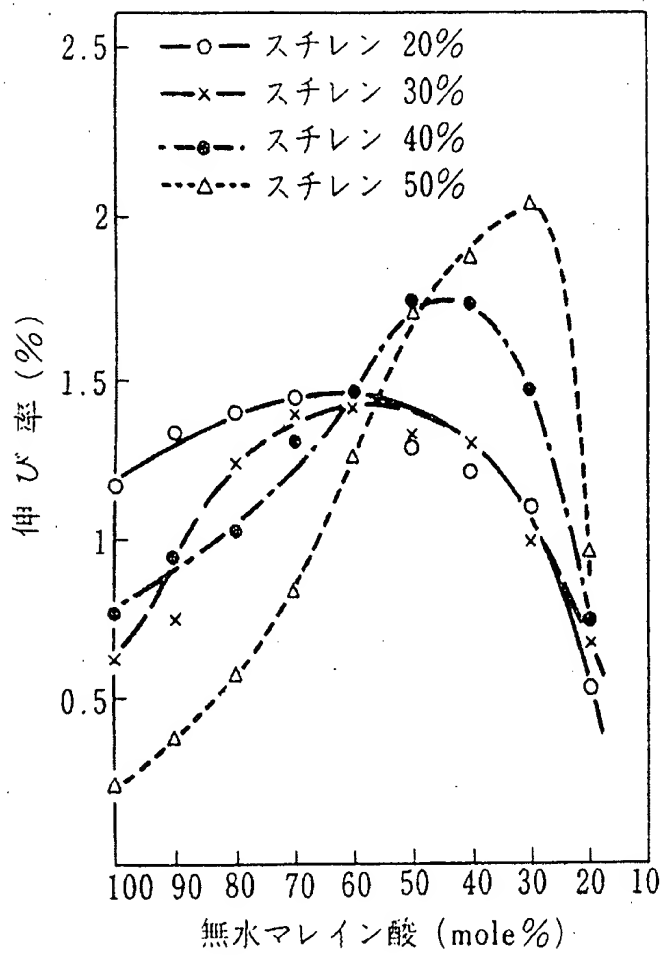

図 7.17　プロピレングリコール～無水フタル酸～無水マレイン酸系樹脂の不飽和度とスチレン量を変化させたときの伸び率

ポリエステル樹脂ハンドブック　　NDC 578

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